REMARKS

The Office action mailed June 18, 2002 has been reviewed. Claims 1 through 17 are pending in this application, of which claims 1 and 7 are independent. Claims 2 through 6 and 8 through 17 are dependent claims. Claims 14 through 17 have been withdrawn from consideration. Upon entry of this Amendment, claims 1, 7, 15, 16 and 17 will have been amended.

1. Claims 1 and 7 have been amended to incorporate the limitation of claim 14. Claim 14 has been canceled without prejudice. Claims 15-17 have been amended to correct improper form noted by the Examiner. Claims 1 and 7 have been amended to more specifically define the invention to which Applicants are entitled.

The claim amendments are made in order to comply with requirements of form or to place the claims in better form for consideration on appeal. As no new matter has been added by these amendments, and good and sufficient reasons have been presented why they are necessary and were not submitted earlier, it is respectfully submitted that they should be entered.

2. Specification

The Examiner objected to the amendment of the Summary of the Invention made March 27, 2002 as introducing new matter. Specifically objected to is the range of mechanical agitation "... at a rate of at least 200 revolutions per minute." Applicants respectfully traverse this objection.

Amendments that merely clarify or make definite that which the originally-filed application either expressly or inherently discloses do not constitute new matter. The amendment to the specification made on March 27, 2002 conformed the Summary of the Invention to amended claim 1. Support for this amendment to the specification and to claim 1 is found in the specification as originally filed. As the Examiner correctly notes, the inventive examples show mechanical agitation at:

- a nominal (i.e., minimal) speed of 500 rpm (Inventive Example 1,
 page 11, lines 19 through 21),
- 200 rpm (Inventive Example 2, page 12, lines 1 through 3) and
- 600 rpm (Comparative Example 3, page 12, lines 15 through 17). There is no indication that these rotational speeds comprise upper limits. As the Examiner correctly notes, the specification at page 4, lines 26-29 states "...200 rpm...provides sufficient shear" (emphasis added). This statement clearly establishes an open-ended range consistent with the phrase "at least 200 revolutions per minute." In view of the examples, it clearly cannot mean an upper limit of 200 rpm and cannot mean no speed other than 200 rpm.

Moreover, this statement must be interpreted in view of the oscillating disk viscosity data of Figure 2 and the discussion at page 6, lines 5 through 16. As shown in Figure 2, as frequency (shear rate) is increased, viscosity of the shear-thinnable fluid drops monotonically without apparent limit.

"With respect to changing numerical range limitations, the analysis must take into account which ranges one skilled in the art would consider inherently supported by the discussion in the original disclosure" M.P.E.P. 2163.05 III. Range Limitations

It is submitted that the aforementioned passages in the original disclosure meet this criterion. Accordingly, reconsideration and entry of the amended Summary of the Invention respectfully is requested.

3. Claim Objections

Claims 14 through 17 have been objected to as being in improper form. Claim 14 has been cancelled without prejudice. Claims 15 through 17 have been amended to depend in the alternative.

4. Claim Rejections 35 USC § 112

Claims 1 through 10 stand rejected under 35 USC § 112, first paragraph. Examiner states that "In the original disclosure applicant does not provide support for agitating at a rate at least 200 rpm as included in claims 1 and 7."

Applicants respectfully request the withdrawal of this rejection for the reasons stated in Paragraph 2 of this Amendment. It is submitted that the aforementioned passages in the original disclosure meet criteria to establish support for the amendment. Accordingly, reconsideration and withdrawal of the rejection respectfully is requested.

5. Claim Rejections 35 USC § 103

Claims 1 through 10 stand rejected under 35 USC 103(a) as being unpatentable over three newly cited references, namely U.S. Patent No. 3,539,326 issued to Otsuka et al. (the Otsuka or 't326 patent), in view of U.S. Patent No. 3,988,398 Frenken et al. (the Frenken or '398 patent) and U.S. Patent No. 3,083,406 issued to Hoogendonk (the Hoogendonk or '406 patent).

Applicants respectfully request withdrawal of this rejection. It is respectfully submitted that the Examiner has failed to make a *prima facie* case of obviousness with respect to this newest combination of references. There must be a basis in the art for combining or modifying the references. Obviousness cannot be established by combining the teachings of the prior art to produce the claimed invention, absent some teaching, suggestion or incentive supporting the combination. There is no suggestion in Otsuka, Frenken or Hoogendonk to achieve the invention for the reasons stated below. The Applicants' disclosure may not be used to hunt through the prior art for the claimed elements and combine them.

Shear-Thinnable Mixture

Claims 1 and 7, as amended, each recite a limitation: (c) reacting said components at a temperature and for a time sufficient to form a shear-thinnable mixture." In contrast, the '326 patent describes nitrogen, phosphorus and/or potassium (NPK) fertilizers wherein the viscosity of the molten materials is increased to the point where fluidity was lost (Examples 1, 2, and 6). Neither is there a teaching in the '398 patent or the '406 patent of the formation of a shear thinnable fluid. The '398 patent also discusses NPK-fertilizers (Col.1, line

42) "...material having high and possibly variable viscosity..." (Col. 1, line 52-53). As illustrated by the '326 patent, this teaching involves materials having viscosities that may increase to the point where fluidity is lost. There is no teaching of, or directed to, shear-thinning fluids.

The'406 patent discusses a "thixotropic" melt (Column 1, line 61). However, the shear-thinning fluid of the invention is one whose viscosity decreases with increased shear rate. This type of fluid is known as a "pseudoplastic" fluid. It is to be distinguished from a thixotropic fluid whose viscosity decreases with time at constant shear rate. (Ullmann's Encyclopedia of Industrial Chemistry, Fifth, Completely Revised Edition, Vol. B1, P.5-26 to 5-28, VCH Verlagsgesellschaft, Weinheim, Germany, 1990). For the convenience of the Examiner, a copy of the relevant pages from this reference is submitted with this Amendment.

It is submitted that the combination of Otsuka, Frenken and Hoogendonk fails to teach the limitation that a shear-thinnable fluid is formed. To establish *prima facie* obviousness of a claimed invention, all claim limitations must be taught, or suggested by the prior art. *In re Royka*, 490 F.2d 981, 180 USPQ 580 (CCPA 1974). All the words in a claim must be considered in judging the patentability of that claim against the prior art. *In re Wilson*, 424 F2.d 1382, 1385, 165 USPQ 494,496 (CCPA 1970). The prior art itself must suggest or teach the claimed invention, and thus the obviousness of making the claimed invention, without the slightest recourse to the teachings of the patent application. Without independent suggestion found in the prior art, the art merely is inviting unguided and speculative experimentation.

Thus, the Examiner has failed to make a *prima facie* case of obviousness with respect to independent claims 1 and 7, as amended, and therefore dependent claims 2 through 6 and 8 through 10. Accordingly, reconsideration of the foregoing rejection respectfully is requested.

Agitation

7, Independent claims 1 and as amended, each d) m chanically agitating said shear-thinnabl mixture at a rotational spe d of at least 200 revolutions per minute in a prill head wherein essentially the entire liquid volume in said prill head is swept by an agitator to shear thin said shear-thinnable mixture. The '326 patent does not employ a prill head. The materials to be prilled are combined in a melt mixing tank 3 and then dropped onto a metal screen, a sieve plate or a wheel having radial bars 16. The Frenken and Hoogendonk patents teach the use of a prill head. Consequently, there is no motivation, either express or implied to combine the teachings of Frenken and/or Hoogendonk, with those of Otsuka.

Moreover, the present invention may utilize either a stationary agitator and a rotating bucket, or a rotating agitator and a stationary bucket. The '398 patent provides a rotating agitator and a rotating bucket. In comparison to the '398 patent therefore, the present invention eliminates an element while retaining the prilling functionality, an indicia of unobviousness.

The Hoogendonk '406 patent does not provide for mechanically agitating the shear-thinnable mixture at a rotational speed of at least 200 revolutions per minute in a prill head wherein essentially the entire liquid volume in said prill head is swept by an agitator.

It is submitted that the combination of Otsuka, Frenken and Hoogendonk fails to teach the step of mechanically agitating said shear-thinnable mixture at a rotational speed of at least 200 revolutions per minute in a prill head wherein essentially the entire liquid volume in said prill head is swept by an agitator to shear thin said shear-thinnable mixture. To establish *prima facie* obviousness of a claimed invention, all claim limitations must be taught or suggested by the prior art. *In re Royka*, 490 F.2d 981, 180 USPQ 580 (CCPA 1974). All the words in a claim must be considered in judging the patentability of that claim against the prior art. *In re Wilson*, 424 F2.d 1382, 1385, 165 USPQ 494,496 (CCPA 1970). The prior art itself must suggest or teach the claimed

invention, and thus the obviousness of making the claimed invention, without the slightest recourse to the teachings of the patent application. Without independent suggestion found in the prior art, the art merely is inviting unguided and speculative experimentation.

Thus, the Examiner has failed to make a *prima facie* case of obviousness with respect to independent claims 1 and 7, as amended, and therefore dependent claims 2 through 6 and 8 through 10. Accordingly, reconsideration of the foregoing rejection respectfully is requested.

6. In light of the foregoing amendment and remarks, it is submitted that independent claims 1 and 7 and dependent claims 2 through 6 and 8 through 10 are allowable and should be passed to issue. Applicants respectfully request the same.

The Examiner is invited to call the undersigned attorney if there are any unresolved issues to discuss same.

This is intended to be a complete response to the Office action mailed June 18, 2002.

Respectfully submitted,

JAMES A. KWEEDER ET AL

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I hereby certify that this correspondence is being deposited with the United States Patent & Trademark Office via facsimile to Examiner Robert A. Madsen, Group Art Unit 1761, at RightFax No. 703-872-9311 (for After Final Communications) on October 9, 2002.

rgaret S. Millikin

IN THE CLAIMS:

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- 1. (Twice Amended) A method to prill a shear-thinnable mixture comprising the steps of:
 - a) providing a molten first component;
 - b) mixing at least a second component with said molten first component;
 - c) reacting said components at a temperature and for a time sufficient to form a shear-thinnable mixture;
 - d) mechanically agitating said shear-thinnable mixture at a rotational speed of at least 200 revolutions per minute in a prill head wherein essentially the entire liquid volume in said prill head is swept by an agitator to shear thin said shear-thinnable mixture; and
 - e) permitting said shear-thinned mixture to flow through holes in said prill head under the influence of a force selected from the group consisting of static pressure and centrifugal force.

- 7. (<u>Twice</u> Amended) A method to prill a shear-thinnable mixture through small prill holes comprising the steps of:
 - a) providing a molten first component;
 - b) mixing at least a second component with said molten first component;
 - c) reacting said components at a temperature and for a time sufficient to form a shear-thinnable mixture;

mechanically agitating said shear-thinnable mixture at a rotational speed of at least 200 revolutions per minute in a prill head wherein essentially the entire liquid volume in said prill head is swept by an agitator to shear thin said shear-thinnable mixture;

- e) wiping the surface of said prill head with surface wiping blades; and
- f) permitting said shear-thinned mixture to flow through small holes in said prill head under the influence of a force selected from the group consisting of static pressure or centrifugal force.
- [14. The prilling method according to claims 1 and 7, wherein essentially the entire liquid volume in said prill head is swept by an agitator.]
- 15. (Amended) The prilling method according to either claim[s] 3 or [and] claim 8, wherein the reaction time is about 10 minutes to about 15 minutes.
- 16. (Amended) The prilling method according to either claim[s] 3 or [and] claim 8, wherein the reaction temperature is at least about 180°C to about 200°C.
- 17. (Amended) The prilling method according to either claim[s] 3 or [and] claim 8, wherein the ammonium nitrate and the ammonium sulfate are present in equimolar amounts.



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The stress is undetermined when $|\tau| < \tau$,. The term y/|y| is necessary to ensure that the stress takes on the same sign as the shear rate and η_{pl} is the plastic viscosity. The true viscosity n approaches η_{pi} only when $\eta_{pi}|\dot{\gamma}|\gg r_{\gamma}$. In contrast the viscosity for the Bingham and any yield stress material becomes unbounded as $|\dot{\gamma}| \rightarrow 0$.

When the shear stress-shear rate function is nonlinear the data are often fit by a Herschel-Buckley model:

$$\tau = \left(\frac{\tau_{y}}{|\dot{\gamma}|} + k \dot{\gamma}^{n-1}\right) \dot{\gamma} \quad \text{for} \quad |\tau| \ge \tau_{y}$$
 (105 a)

$$\eta = \frac{\tau_y}{|\dot{y}|} + k|\dot{y}|^{n-1} \quad \text{for} \quad |\tau| \ge \tau, \tag{105 b}$$

The parameters k and n are obtained by plotting $\tau - \tau_y$ versus y on logarithmic coordinates. Another equation often used to describe yield stress materials is the Casson equation

$$\tau = \left(\frac{\tau_y^{1/2}}{|\hat{y}|^{1/2}} + \eta_p^{1/2}\right)^2 \hat{y} \quad \text{for} \quad |\tau| \ge \tau, \quad (106 \, a)$$

$$\eta = \left(\frac{\tau_y^{1/2}}{|\dot{y}|^{1/2}} + \eta_{pl}^{1/2}\right)^2 \quad \text{for} \quad |\tau| \ge \tau, \tag{106b}$$

Typical shear stress-shear rate data for a meat extract concentrate and for a tomato soup concentrate are presented in Figures 24 and 25, respectively. The meat extract behaves like a Bingham solid while the tomato soup concentrate exhibits Herschel-Buckley behavior.

The yield stress is associated with a three-dimensional structure that must deform elastically before flow can occur. It is commonly observed in fine particle suspension systems, especially at

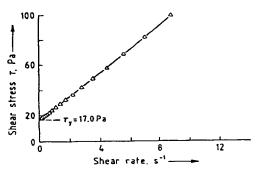


Figure 24. Shear stress-shear rate data (flow curve) for a meat extract

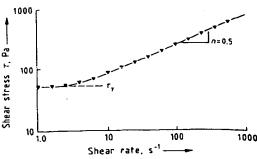


Figure 25. Shear stress-shear rate data (flow curve) for a tomato soup concentrate

high particle concentrations. Common examples are toothpaste, paint, oil well drilling muds, foams, and many food and pharmaceutical products.

4.1.2. Shear-Thinning (Pseudoplastic) Fluids

Solutions and melts of flexible macromolecules and many suspensions exhibit pseudoplastic shear stress-shear rate behavior such as that illustrated in Figure 23 and shown specifically for a polymer solution in Figure 26. Here the ratio $\tau/\dot{\gamma}$ (i.e., η) is a decreasing function of $\dot{\gamma}$. Such a fluid has a shear-thinning viscosity and is referred to as pseudoplastic. The decrease of viscosity with increasing rate of deformation can usually be attributed to the breakdown of a structure at the colloidal or molecular level. Macromolecules will become more aligned and hence less entangled and less resistant to deformation as the deformation rate is increased.

The viscosity of pseudoplastic fluids is typically plotted versus shear rate on logarithmic coordinates because the viscosity for such fluids can change by orders of magnitude over several decades of shear rate. The data for the polyacrylamide solution shown in Figure 26 are typical for a macromolecular fluid. The viscosity approaches a constant value at low shear rates called the zero-shear viscosity η_0 and a constant value in the limit of very high shear rates called the infinite shear viscosity n. Experimental measurements in both regions can be difficult, particularly in the infinite shear region. Thus data are often only available between the two limits. Between the two limits the shear stress (or viscosity)

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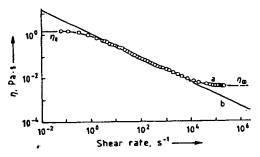


Figure 26. Viscosity as a function of shear rate for a polyacrylamide solution

is often linear with shear rate over several decades on logarithmic coordinates. In this restricted region

$$\log \tau = \log k + n \log \gamma \tag{107a}$$

$$\log \eta = \log k + (n-1)\log \dot{\gamma} \tag{107b}$$

where n is the slope in the linear region of the plot (n = 0.41) for the data in Fig. 26). Thus

$$\tau = k|\dot{\gamma}|^{n-1}\dot{\gamma} \tag{108a}$$

$$\eta = k |\dot{\gamma}|^{n-1} \tag{108b}$$

Equation (108) is known as the power law and sometimes as the Ostwald—de Waele model. This law is empirical and must fail for both high and low shear rates. The factor k is very temperature sensitive (and concentration sensitive for suspensions), but n is typically insensitive to temperature (and concentration) changes.

Many empirical and semi-empirical equations have been proposed to represent the viscosity data for shear-thinning fluids. The Meter model:

$$\eta = \eta_{\infty} + \frac{\eta_0 - \eta_{\infty}}{1 + \left| \frac{\tau}{\tau_{1/2}} \right|^{\alpha - 1}} \tag{109}$$

has four parameters, η_0 , η_{∞} , $\tau_{1/2}$, and α with the following asymptotic behavior

$$\frac{\tau}{\tau_{1/2}} \to 0, \quad \eta \approx \eta_0 \tag{109a}$$

$$\frac{\tau}{\tau_{\rm cm}} \to \infty, \quad \eta \approx \eta_{\infty}$$
 (109 b)

and for

$$1 \ll \left| \frac{\tau}{\tau_{1/2}} \right|^{\alpha - 1} \ll \frac{\eta_0}{\eta_\infty}$$

then

$$\eta = \frac{\eta_0 - \eta_{\infty}}{\tau_{1/2}^{1-\alpha}} |\tau|^{1-\alpha} \tag{109 c}$$

The Meter model exhibits power-law behavior in the intermediate region, with $\alpha = 1/n$; $\tau_{1/2}$ is the stress at which the viscosity is midway between the values of η_0 and η_{∞} . If η_{∞} is taken to be zero, which is often a good approximation for molten polymers, then $\tau_{1/2}$ is the stress at which the viscosity drops to one-half the zero-shear value. In this case Equation (109) is known as the Ellis model. Line (a) through the data in Figure 26 is the fit of the Meter model to the data with $\eta_0 = 1.43 \text{ Pa s}, \ \eta_\infty = 0.004 \text{ Pa s}, \ \tau_{1/2} = 1.21 \text{ Pa},$ and $\alpha = 2.43$. The power-law fit is line (b) with n = 0.41 (= $1/\alpha$). The Ellis model follows the Meter model at low shear rates (incorporating the zero-shear viscosity behavior) and the power-law at higher shear rates.

4.1.3. Shear-Thickening (Dilatant) Fluids

Some suspensions with a high concentration of solids and with a relatively uniform particlesize distribution have a viscosity that increases with shear rate. An example for a titanium dioxide suspension is shown in Figure 27 where at the

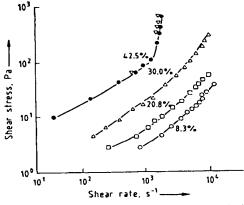


Figure 27. Shear stress-shear rate for a titanium dioxide suspension [20]

highest volume fraction of solids (42.5%) the viscosity increases with shear rate beyond a critical shear rate. The increase in stress observed at this point is related to a shear-induced structure. The phenomena is easily illustrated in the kitchen by mixing a concentrated suspension of corn starch and water. Data for shear-thickening liquids are usually fit by equations such as those for shear-thinning fluids but with $\eta_{\infty} = 0$ and n > 1 (or $\alpha < 1$). Dramatic equipment failures have been documented in the processing industries where a dilatant fluid response was not anticipated.

4.1.4. Fluids with Time-Dependent Viscosity (Thixotropic Fluids)

A special nomenclature is used in cases when the viscosity varies with the time of shear and the shear rate. A material whose apparent viscosity decreases with the time of shear is said to be thixotropic and one whose viscosity increases with time of shear is said to be rheopectic. The behavior of these materials depends not only on the time of shear but also on the past shear and thermal history. Rheopectic fluids are encountered very rarely and hence the discussion in this section will be confined to thixotropic fluids.

If the flow curve is measured in a single experiment in which the shear rate is steadily increased at a constant rate from zero to some maximum value and then decreased at the same rate to zero shear rate, a form of a hysteresis loop as is shown in Figure 28 is observed. The position of this loop, its shape, and the area within the

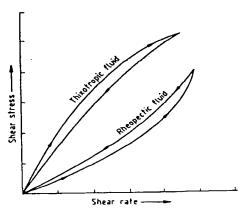


Figure 28. Time dependent fluids

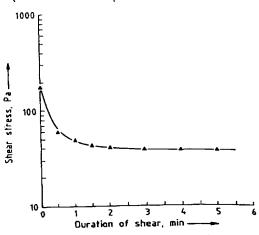


Figure 29. Shear stress versus time of shear for an Australian waxy crude oil at 10 °C and a shear rate of 458 s-1

loop depends on the rate at which the shear rate is increased and decreased as well as the past thermal and shear history of the material. This type of measurement has little meaning for the solution of practical flow problems. What is required is a series of experimentally determined shear stress-shear rate-time curves which can then be converted to a series of flow curves with time of shear as the parameter. Because of the dependence of these curves on shear and thermal history it is important that the curves represent the behavior of a sample of material which has experienced a history approximately the same as that anticipated for the fluid whose flow behavior is to be analyzed.

The difference between thixotropy and pseudoplasticity is thought to be the time element in structural breakdown, which is finite and measurable for the thixotropic fluid and in contrast very small and undetectable for the pseudoplastic fluid. A practical example of time-dependent behavior is the waxy crude oils which in addition also exhibit a yield stress (see Fig. 29). It should be noted that a fluid can show time-dependent and viscoelastic behavior (for viscoelastic behavior see Section 4.3). Some liquid crystal polymers appear to be rheopectic and viscoelastic.

4.2. Fully Developed Tube Flow

The flow in tubes (pipes) of circular cross section is the flow field of the greatest interest.